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The view, opinions, and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A brief description, including bibliography and personnel, is given of 26 years of ARO support in the area of photophysical processes and molecular transformations of excited states is given.

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FINAL REPORT ON U.S. ARMY SPONSORED RESEARCH

by Professor Howard E. Zimmerman

University of Wisconsin, Madison, Wisconsin

December 14, 1981

To the U. S. ARMY RESEARCH OFFICE

Grant DAAG29 78 G 0204

Approved for Public Release

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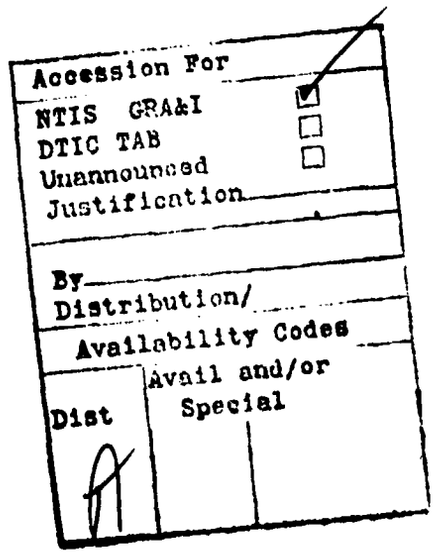

1. Period of Support by U. S. Army Research Office:

The U. S. Army Research Office supported our research in a series of grants beginning in September 1956 and ending in September of 1981.

The most recent grant covered the period September 25, 1978 through September 24, 1981.

This final report formally covers this last three-year period. However, in view of the very extended support of our research by the U.S. Army Research Office, and the cessation of this support after a 25-year period, it seems appropriate to cover the entire period to some extent.

This report is slightly delayed due to lack of secretarial assistance deriving from the precipitous and unexpected cessation of ARO support. Thus the report has had to be prepared in its entirety by the Principal Investigator himself.

2. Most Recent Grant Number: DAAG29-78-G-0204.
Considering the very long period of support, our records do not maintain the various grant numbers over the entire period of 25 years.

3. Title of Research Project: Our most recent title is: "Study of Photophysical Processes and Molecular Transformations of Excited States."

The title previous to this one was "A Study of Unstable Organic Species and Unusual Organic Transformations."

The first title covers the last six years of research support.

4. Personnel Supported During the Last 3 Year Period:

Individual and Appt	Present Position
Jeffrey Blood (Res Asst)	Completed Ph.D., now at Eastman Kodak Co.
Richard Bunce (Res Asst)	Completed Ph.D., now Postdoctoral Fellow at Berkeley
Craig Caufield (Res Asst)	Still doing graduate study.
Paula Clausen (Secretary)	Moved to another position.

Thomas Gannett (Res Asst)	Completed Ph.D.,now at E. I. DuPont de Nemours and Co.
Timothy Hirzel (Res Asst)	Completed Ph.D., now at Monsanto, St. Louis
Doreen Lynch (Research Asst)	Still doing graduate study.
John Nuss (Res Asst)	Still doing graduate study.
Robert Pasteris (Res Asst)	Completed Ph.D., now at E. I. DuPont de Nemours and Co.
John Penn (Res Assistant)	Completed Ph.D., now Postdoctoral Fellow at Univ. of Utah
Richard Swafford (Res Asst)	Still doing graduate study.
Anthony Tantillo (Res Asst)	Still doing graduate study.

5. Publications of ARO Supported Research

a. A complete list of publications describing A.R.O. supported research is attached.

b. Publications during the most recent, three year grant period as listed in the following. These total 17 in number.

"Stereochemistry in the Di-π-Methane Rearrangement; Aryl Migration. Exploratory and Mechanistic Organic Photochemistry," H. E. Zimmerman, T. P. Gannett and G. E. Keck, J. Amer. Chem. Soc., 1978, 100, 323-324.

"Unsual Regioselectivity in the Di-π-Methane Rearrangement. Reaction Inhibition and Control by Electron Donating Substituents. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman and W. T. Gruenbaum, J. Org. Chem., 1978, 43, 1997-2005.

"Vinylcyclopropene Photochemistry; Photochemistry Applied to Organic Synthesis. Exploratory and Mechanistic Organic Photochemistry," H. E. Zimmerman and S. M. Aasen, J. Org. Chem., 1978, 43, 1493-1506.

"Differing Substituent Effects on Rates and Regioselectivities in the Di-π-Methane Rearrangement. New Photochemical Theory: Excitation Distribution (the ΔP Matrix), Reaction Inhibition by Vertical Excited State Stabilization, Ground State vs. Excited State Control," H. E. Zimmerman, W. T. Gruenbaum, R. T. Klun, M. G. Steinmetz, and T. R. Welter, J.C.S. Chemical Communications, 1979, 228-230.

"Control of Regioselectivity and Excited State Singlet Reaction Rates by Substitution in the Di-π-Methane Rearrangement. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman and T. R. Welter, J. Amer. Chem. Soc., 1978, 100, 4131-4145.

"The Aryl-Vinyl Methane Version of the Di-π-Methane Rearrangement. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman, M. G. Steinmetz and C. L. Kreil, J. Amer. Chem. Soc., 1978, 100, 4146-4162.

"The Di-π-Methane Rearrangement of Systems With Simple Vinyl Moieties. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman and R. T. Klun, Tetrahedron, 1978, 45, 1775-1803.

"Molecular Control of Excited State Cross-Conjugated Triene Rearrangements. Exploratory and Mechanistic Organic Photochemistry," H. E. Zimmerman and D. R. Diehl, J. Amer. Chem. Soc., 1979, 101, 1841-1857.

"Stereochemistry of the Aryl-Vinyl Version of the Di-π-Methane Rearrangement," H. E. Zimmerman, T. P. Gannett and G. E. Keck, J. Org. Chem., 1979, 44, 1982-1989.

"Unusual Organic Photochemistry Effected by Cyano and Methoxy Substitution. Exploratory and Mechanistic Organic Photochemistry," H. E. Zimmerman, D. Armesto, M. G. Amezua, T. P. Gannett, and R. P. Johnson, J. Amer. Chem. Soc., 1979, 101, 6367-6383.

"The Bicycle Rearrangement: Relationship to the Di-π-Methane Rearrangement and Control by Bifunnel Distortion. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman and R. E. Factor, J. Amer. Chem. Soc., 1980, 102, 3538-3548.

"Rod-like Organic Molecules; Energy Transfer Studies Using Single Photon Counting," H. E. Zimmerman, T. D. Goldman, T. K. Hirzel and S. P. Schmidt, J. Org. Chem., 1980, 45, 3933-3951.

"Regioselectivity in Cyclohexadienone Photochemistry; the Role of Zwitterions in Type A Photochemical and Dark Rearrangements. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman and R. J. Pasteris, J. Org. Chem., 1980, 45, 4864-4875.

"Type A Zwitterions and Cyclohexadienone Photochemical Rearrangements. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman and R. J. Pasteris, J. Org. Chem., 1980, 45, 4876-4891.

"Di-π-Methane Hypersurfaces and Reactivity; Multiplicity and Regioselectivity; Relationship Between the Di-π-Methane and Bicycle Rearrangements," H. E. Zimmerman and R. E. Factor, Tetrahedron, 1981, 37, Supplement 1, 125-141.

"New Reactions and Theory in Organic Photochemistry: The 1,3-Vinyl Migration and its Relevance to Exchange Integral Control," H. E. Zimmerman, J. H. Penn and M. R. Johnson, Proc. Natl. Acad. Sci. USA, 1981, 78, 2021-2025.

"Evaluation of Single Photon Counting Measurements of Excited State Lifetimes," H. E. Zimmerman, J. H. Penn and C. W. Carpenter, Proc. Natl. Acad. Sci. USA, scheduled for January or February.

6. Technical Details of Research During the Three Year Period. A comprehensive report of accomplishments was made in our recent renewal application, namely the renewal which was turned down. Additionally, detailed semiannual technical reports have been turned in.

7. General Accomplishments During the Three Year Period. Our research has been especially successful in adding to basic research knowledge of the photophysical and photochemical processes available to and utilized by excited state molecules.

8. Accomplishments Made Possible by the 26 Years of ARO Research Support:

Our ARO grant support began in 1956 when the Principal Investigator, Howard E. Zimmerman, was a young Assistant Professor. The impact of ARO support has been three-fold. On one hand it has contributed a number of basic and important scientific findings and chemical concepts to the field. Secondly, it has contributed to the development of the scientific recognition of the Principal Investigator. Finally, it has supported the graduate and postdoctoral studies of a large number of very highly skilled and creative students. Many of these have gone on into independent careers and are now known professionally. Thus 66 former students of Howard E. Zimmerman's former students have become professors, and an appreciably large fraction of these were supported by ARO. This has created considerable good will for ARO and its objectives.

The Principal Investigator received the 1976 James Flack Norris Award of the American Chemical Society, the 1979 Halpern Award for Photochemistry of the New York Academy of Science, and was elected in 1980 to the National Academy of Sciences. It was, indeed, ARO support which helped make this recognition possible.

Scientifically, ARO has had major impact.

Thus, even the first paper describing ARO sponsored research, Reference 10 in the complete bibliography below is now quoted as laying the groundwork for the stereochemistry of aldolization. This paper provided evidence for a six-membered transition state in aldolization, Reformatsky and related reactions.

The photochemical papers provided basic theory which allowed and catalyzed the rapid growth of photochemical research. The Principal Investigator's research group was one of the two original groups in the U.S. which initiated detailed photochemical studies. Most photochemical research now follows in these footsteps. Note the photochemical references in the complete bibliography below.

Reference in the complete bibliography describes the Möbius-Hückel Concept which has become an integral part of modern chemistry. It is to be found now in most sophomore organic chemistry textbooks.

The above are some salient examples of the results of the 26 years of ARO research.

9. General Comments About ARO Funding of Basic Research:

It is obvious that the Principal Investigator appreciates the 26 years of support of his research by ARO. This support has generated very warm feelings towards ARO also by students supported in their Ph.D. and Postdoctoral endeavors. Beyond this, fellow students supported by other means clearly developed positive feelings about ARO in the process. Many of these people now are men of international stature in research.

However, ARO's precipitous withdrawal of support from the Principal Investigator was at a time when his research has been most successful and when ARO's confidence in his efforts should have been most clearly justified. This withdrawal at a time when research funding is most difficult, caused severe hardship. While the Principal Investigator recognized the ever increased demand for relevancy, he was assured by Army researchers that relevance was not lacking in his work. Also he is a bit baffled by support of younger people of lesser stature in the same period as ARO's withdrawal. That some of this support is for photochemical research pioneered by the Principal Investigator is not too understandable.

Nevertheless, in light of the 26 years of ARO loyal support, this present comment reflects primarily bafflement. ARO's past help, encouragement, understanding and loyalty won't be forgotten.

10. Examples of the Principal Investigator's Former Students, ARO Supported and Not, Benefitting From ARO Support:

Professor Albert Padwa - Emory University. Presently a leading Organic Chemist.

Professor Harry H. Morrison - Purdue University. A leading photochemist.

Professor David I. Schuster - New York University. Also a leading photochemist.

Professor Jaques Nasielski - Head of the Chem. Dept., Univ. of Brussels and an internationally known chemist.

Professor John S. Swenton - Ohio State Univ. A leading organic chemist.

Professor Hiizu Iwamura - Institute for Molecular Science, Japan. One of the most influential younger Japanese photochemists.

Professor Gary Keck - University of Utah. Said to be the most promising and brilliant young organic chemist.

Professor Patrick S. Mariano - University of Maryland. Another of the country's most energetic and productive organic chemists.

Professor John J. McCullough - McMaster University. One of Canada's most outstanding few organic photochemists.

Professor James Pincock - Dalhousie University. A particularly energetic and brilliant young Canadian photochemist.

Professor Albert C. Pratt - Dublin. The most eminent Irish organic photochemist.

Professor Laren Tolbert - University of Kentucky. Another well known young organic photochemist.

11. List of ARO Supported Personnel and Current Position

BARCKSTROM, Peter Assistant Professor	Department of Organic Chemistry The Royal Institute of Technology S-100 44 Stockholm 70, SWEDEN
BAKER, Joseph T.	Roche Research Institute of Marine Pharmacology 4-10 Inman Road, Dee Why P.O. Box 255, Dee Why, 2099 N.W.W., AUSTRALIA
CRUMRINE, David Associate Professor	Department of Chemistry Loyola University of Chicago Chicago, Illinois 60626

EBERBACH, Wolfgang Professor (C2)	Chemisches Laboratorium der Universität Albertstr. 21 7800 Freiburg WEST GERMANY
FLECHTNER, Thomas Associate Professor	Department of Chemistry Cleveland State University Cleveland, Ohio 44115
HORNBACK, Joseph Associate Professor	Department of Chemistry University of Denver Denver, Colorado 80210
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MARIANO, Patrick S. Professor	Department of Chemistry University of Maryland College Park, Maryland 20742
MC CULLOUGH, John J. Professor	Department of Chemistry McMaster University Hamilton, Ontario CANADA
MC KELVEY, Ronald Academic Staff	Department of Chemistry University of Wisconisin-LaCross LaCrosse, Wisconsin 54601
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University of Utah
Salt Lake City, Utah

PINCOCK, James
Associate Professor
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Dalhousie University
Halifax, Nova Scotia, CANADA

PRATT, Albert
Professor
School of Chemical Sciences
National Institute for Higher Education
Dublin, IRELAND

TOLBERT, Laren
Associate Professor
Department of Chemistry
University of Kentucky
Lexington, Kentucky

VIRIOT, Marie-Laure
Assistant Professor
INPL (Ensic)
Laboratoire De Recherches De
Chemie Generale
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12. Full List of Publications by H. E. Zimmerman Utilizing A.R.O Support:

10. "The Stereochemistry of the Ivanov and Reformatsky Reactions. I," H. E. Zimmerman and M. D. Traxler, J. Amer. Chem. Soc., 1957, 79, 1920-1923.

16. "Overlap Control of Carbanionoid Reactions. I. Stereoselectivity in Alkaline Epoxidation," H. E. Zimmerman, L. Singer and B. S. Thyagarajan, J. Amer. Chem. Soc., 1959, 81, 108-116.

17. "Overlap Control of Carbanionoid Reactions. II. Stereochemistry of the Perkin and Related Condensation Reactions," H. E. Zimmerman and L. Ahramjian, J. Amer. Chem. Soc., 1959, 81, 2086-2091.

23. "Overlap Control of Organic Reactions. III. The Stereochemistry of the Darzens Reaction," H. E. Zimmerman and L. Ahramjian, J. Amer. Chem. Soc., 1960, 82, 5459-5466.

28. "Overlap Control of Organic Reactions. III. The Stereochemistry of the Darzens Reaction," H. E. Zimmerman and L. Ahramjian, J. Amer. Chem. Soc., 1960, 82, 5459-5466.

36. "A General Theory of Photochemical Reactions. VII. Mechanisms of Epoxy Ketone Reactions," H. E. Zimmerman, B. R. Cowley, C-Y. Tsang, and J. W. Wilson, J. Amer. Chem. Soc., 1964, 86, 947-948.

39. "A Study of Hindered Divalent Carbon Species and Diazo Compounds," H. E. Zimmerman and D. H. Paskovich, J. Amer. Chem. Soc., 1964, 86, 2149-2160.

40. "Photochemical Reactions of 2,3-Epoxy-2-methyl-3-phenylindanone," H. E. Zimmerman and R. D. Simkin, Tetrahedron Lett., 1964, 1847-1851.

41. "The Photochemical Reaction of Benzoquinone With Tolan," H. E. Zimmerman and L. Craft, Tetrahedron Lett., 1964, 2131-2136.

59. "Chronology in Photochemical Mechanisms. The Reaction of 6-Phenyl-6-p-Cyanophenylbicyclo[3.1.0]hex-3-en-2-one. Mechanistic Organic Photochemistry. XXV," H. E. Zimmerman and J. O. Grunewald, J. Amer. Chem. Soc., 1967, 89, 3354-3356.

60. "Mechanistic Organic Photochemistry. XXIV. The Mechanism of the Conversion of Barrelene to Semibullvalene. A General Photochemical Process," H. E. Zimmerman, R. W. Binkley, R. S. Givens, and M. A. Sherwin, J. Amer. Chem. Soc., 1967, 89, 3932-3933.

61. "Pathways Leading From Excited-State Reactant to Ground-State Products in Dienone Photochemistry. Mechanistic and Exploratory Organic Photochemistry. XXVI," H. E. Zimmerman and J. O. Grunewald, J. Amer. Chem. Soc., 1967, 89, 5163-5172.

63. "π-π^* Rearrangements. The Photochemistry of 1-Methylene-4,4-Diphenyl-2,5-cyclohexadiene as a Dienone Analog. Mechanistic and Exploratory Organic Photochemistry. XXIX," H. E. Zimmerman, P. Hackett, D. F. Juers, and B. Schröder, J. Amer. Chem. Soc., 1967 89, 5973-5974.

71. "The Stereochemistry and Mechanism of the Photochemical Interconversion of cis- and trans-5,6-Diphenylbicyclo[3.1.0]hexan-2-ones. Mechanistic Organic Photochemistry. XXXII," H. E. Zimmerman, K. G. Hancock, and G. Licke, J. Amer. Chem. Soc., 1968, 90, 4892-4911.

78. "Relative Rates of Aryl Migrations in Excited State Transformations. Mechanistic and Exploratory Organic Photochemistry. XXXIX," H. E. Zimmerman and N. Lewin, J. Amer. Chem. Soc., 1969, 91, 879-886.

79. "Energy Barriers in Photochemical Molecular Rearrangements. Mechanistic and Exploratory Organic Photochemistry. XL. H. E. Zimmerman and W. R. Elser, J. Amer. Chem. Soc., 1969, 91, 887-896.

80. "The Di-π-Methane Rearrangement. Interaction of Electronically Excited Vinyl Chromophores. Mechanistic and Exploratory Organic Photochemistry. XLI," H. E. Zimmerman and P. S. Mariano, J. Amer. Chem. Soc., 1969, 91, 1718-1727.

81. "Synthesis and Physical Properties of Barrelene, a Unique Möbius-like Molecule," H. E. Zimmerman, R. M. Paufler, and M. A. Sherwin, J. Amer. Chem. Soc., 1969, 91, 2330-2338.

82. "The Barrelene to Semibullvalene Transformation. Correlation of Excited State Potential Energy Surfaces With Reactivity. Mechanistic and Exploratory Organic Photochemistry. XLIV," H. E. Zimmerman, R. W. Binkley, R. S. Givens, G. L.Grunewald, and M. A. Sherwin, J. Amer. Chem. Soc., 1969, 91, 3316-3323.

85. "C_8H_8 Interconversions. An Unusual Rearrangement Providing a New Route to Semibullvalene," H. E. Zimmerman, J. D. Robbins and J. Schantl, J. Amer. Chem. Soc., 1969, 91, 5878-5879.

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88. "Organic Photochemistry. LIV. Concertedness, Stereochemistry, and Energy Dissipation in the Di-π-Methane Rearrangement. Source of Singlet-Triplet Reactivity Differences," H. E. Zimmerman and A. C. Pratt, J. Amer. Chem. Soc., 1970, 92, 1409-1411.

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97. "Stereochemical Aspects pf the Di-π-Methane Rearrangement; Mechanistic and Exploratory Organic Photochemistry. LVII," H. E. Zimmerman and A. C. Pratt, J. Amer. Chem. Soc., 1970, 92, 6267-6271.

98. "Electron Delocalization in Molecules Containing Formally Orthogonal π-systems. The Synthesis of 2,4,6-2,',4,',6'-Hexa-t-butyldiphenylacetylene and a Study of Its Radical Anion and Dianion," H. E. Zimmerman and J. R. Dodd, J. Amer. Chem. Soc., 1970, 92, 6507-6515.

99. "Singlet-Triplet Differentiation. Mechanistic and Exploratory Organic Photochemistry. LIX," H. E. Zimmerman and V. J. Hull, J. Amer. Chem. Soc., 1970, 92, 6515-6520.

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101. "Vinylcyclopropane Photochemistry. Mechanistic and Exploratory Organic Photochemistry. LXI," H. E. Zimmerman and T. W. Flechtner, J. Amer. Chem. Soc., 1970, 92, 7178-7183.

102. "Electron and Energy Transfer Between Bicyclo[3.1.0]octane Bridgehead Moieties," H. E. Zimmerman and R. D. McKelvey,, J. Amer. Chem. Soc., 1971, 93, 3638-3645.

104. "Competitive Photochemical Pathways in the Di-π-Methane Rearrangement. Exploratory and Mechanistic Organic Photochemistry. LXIII," H. E. Zimmerman, P. Hackett, D. F. Juers, J. M. McCall, and B. Schröder, J. Amer. Chem. Soc., 1971, 93, 3653-3662.

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106. "The Möbius-Hückel Concept in Organic Chemistry. Application to Organic Molecules and Reactions," H. E. Zimmerman, Accounts of Chem. Res., 1971, 4, 272-280.

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128. "Superfast Singlet Excited State Di-π-Methane Rearrangement and Decay Rates; Picosecond Range Measurements by Single Photon Counting and Use of Magic Multipliers. Mechanistic Organic Photochemistry. LXXXI," H. E. Zimmerman, D. P. Werthemann, and K. S. Kamm, J. Amer. Chem. Soc., 1973, 95, 5904-5095.

130. "A Photochemical 1,3-Sigmatropic Vinyl Migration. Mechanistic and Exploratory Organic Photochemistry. LXXXII," H. E. Zimmerman, D. W. Kurtz, and L. M. Tolbert, J. Amer. Chem. Soc., 1973, 95, 8210-8212.

131. "Single Photon Counting and Magic Multipliers in Direct Measurement of Singlet Excited State Di-π-Methane Rearrangement Rates in the Picosecond Range. Mechanistic Organic Photochemistry. LXXXIII," H. E. Zimmerman, D. P. Werthemann, and K. S. Kamm, J. Amer. Chem. Soc., 1974, 96, 439-449.

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133. "Stereochemistry of the Di-π-Methane Rearrangement at the Methane Carbon. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman, J. D. Robbins, R. D. McKelvey, C. J. Samuel, and L. R. Sousa, J. Amer. Chem. Soc., 1974, 96, 1974-1976.

134. "Stereochemistry as a Probe for Photochemical Reaction Mechanisms," H. E. Zimmerman, Tetrahedron, Van't Hoff - Le Bel Memorial Issue, 1974, 30, 1617-1628.

136. "Stereochemistry at the Methane Carbon in the Di-π-Methane Rearrangement. Mechanistic and Exploratory Organic Photochemistry," H. E. Zimmerman, J. D. Robbins, R. D. McKelvey, C. J. Samuel, and L. R. Sousa, J. Amer. Chem. Soc., 1974, 96, 4630-4643.

141. "A Photochemical Rearrangement Without Light. Dioxetane Effected Phothchemistry by Direct Intramolecular Excitation," H. E. Zimmerman and G. E. Keck, J. Amer. Chem. Soc., 1975, 97, 3527-3528.

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